Photoexcitation and Decay of Hollow Lithium States by Electron Spectroscopy

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A hollow atomic system is an atom or an ion in which at least one inner-shell is empty. Lithium, with one additional electron outside the $1s^2$ core, is the simplest open-shell many electron system. Simultaneous excitation of all three electrons can create hollow atoms of the type nln'l'n"l", with $n \geq 2$. Because all the electrons have $n > 2$ outside an empty K-shell, the role of electron correlations is expected to be very important and so hollow Li constitutes an ideal test case for many-body theory. The decay mechanisms for hollow atomic states are of fundamental interest and also because they may provide insight into physical processes relevant to interaction of hollow atoms with surfaces in ion-surface collisions.

The exploration in *photoabsorption* of the lowest $2s^2 2p\ ^2P^o$ resonance at 142.3 eV energy was first achieved.[1] Soon after, two studies[2,3] using the total photoion yield technique revealed the existence of numerous high lying resonances decaying into Li^+ and Li^{++} final states, in agreement with the predictions of a R-Matrix calculation by L. VoKy.[4] However, neither photoabsorption nor ion yield measurements can discriminate against transitions into various continua of the Li^+ ion, and so cannot test the many-body calculations in detail.

Photoelectron spectroscopy is the only experimental method allowing to determine partial photoionization cross sections to leave the singly charged ion in its different final states. When lithium atoms are photoionized in the 1s shell, according to $1s^2 2s\ ^2S_{1/2} + h\nu \rightarrow 1snl\ ^{1,3}L + el'$, the residual Li^+ ion can be left in any of the following ionic states: $1s2s\ ^3S$ and ^1S (main lines in a photoelectron spectrum, corresponding to single photoionization); $1s2p\ ^3P$ and ^1P, $1s3l\ ^{1,3}L$, and $1snl\ ^{1,3}L$, with $n > 3$ (corresponding to various types of correlation satellites).

In our first measurements at the Super ACO storage ring in Orsay,[4,5] the resolution was modest (0.5 eV and 0.23 eV on the bending magnet and undulator beam lines, respectively.) During the summer of 1995 and 1996, we had access to the 9.0.1 undulator beam line of the Advanced Light Source (A total of 2x20 shifts were used).

We have first made a comparison of the actual photon flux and resolution we obtained with our electron spectrometer (we transferred all our experimental set up from Orsay to Berkeley), i. e., using exactly the same apparatus on all beam lines.[5] Compared to the resolution we had in Orsay, we measured at ALS an improvement in the spectral resolution effectively used to be able to do the experiments of at least a factor 10. We have been able to run the experiment, i.e., to get effectively data within the allocated periods of beam time, with a 40 meV resolution (a factor 6 better than on the undulator at Super ACO), and, in some special cases, with 20 meV. We would have liked to work always with this 20 meV resolution, but the time was too short to do so. At the same time, the photon flux (with 40 meV resolution and also with a resolution on the electron spectrometer improved by a factor of 2) was higher than in Orsay (with a 230 meV resolution) by a factor of 2 to 3. To conclude with this experimental aspect, ALS has provided:

- A much higher spectral resolution
- The capability to use a better resolution of the electron spectrometer
- A higher flux (by a factor 2) even with these improved spectral and experimental resolutions, allowing to have a greater sensitivity in the detection of the electrons.

We briefly summarize the data that we have obtained in 1995 and 1996 which have been published in 1996 and in 1997, or are going to be published:[5-11]

1) We have measured partial photoionization cross sections over the energy range from 142 eV to 164 eV with unprecedented resolution.[5,6] Our measurements are compared with the R-matrix calculations of L. VoKy. There is an amazing agreement between tthe results of our calculations using the R-matrix theory and our experiment, on a relative basis, which confirm the quality of the experimental data and the advance in theory made possible by VoKy. Thanks to the high brightness of ALS, it is the first time that a many-body calculation is tested up to the finest details.

2) We have measured in detail the autoionization of a very weak resonance, the $2p^3$ hollow state at 148.75 eV. Although the cross section is very small, it is a particularly interesting resonance, because the three excited electrons are in the same orbitals (same principal and orbital quantum numbers). This state allows a severe test of the treatment of electron correlations.[7]

3) We have observed a number of Auger lines corresponding to two-step autoionization into the continuum of Li^+ 1s ion. Since many highly excited states of Li^+, such as $2s^2\ ^1S$, $2s2p\ ^3P$, $2p^2$, ...,.are lying very near some hollow atomic states, these states of neutral lithium are sometimes a few tenths of an electron-volt above the highly excited states of Li^+., in such a way that the propability to first decay by autoionization to these Li^+ states is very high. The corresponding electrons have a very low kinetic energy and we do not observe them. But the nln'l' states of Li^+ further decay, in a second step, into the continuum of Li^+, producing Auger electrons with characteristic energies. We observed several of them, some of them for the first time, some others remain still unidentified.[8]

4) We have measured with the highest resolution the autoionization of the $2s^2 2p\ ^2P$ state at 142.31 eV into the $1s2p\ ^1P$ final ionic state. R-matrix calculations predicted this resonance to be quite narrow and the resonant enhancement to be quite large. Our experimental results are in good agreement with the calculations. They also allow us to measure an acurrate value for the natural width of this state. The value we obtained, < 0.12 eV, is lower than all previous experimental determinations. It is an important parameter for the description of these hollow states.[5,6]

5) We have succeded to excite hollow states from the laser-excited $1s^2 2p$ lithium atoms.[9] The two main resonances predicted by R-matrix calculations to be near 143 eV ($2s2p^2\ ^2S$) and 145 eV ($2s2p^2\ ^2D$) have been measured at these energies and partial cross sections have been measured for photoionization into the ($1s2p\ ^1P$ + and ^3P + el channels. We have also measured higher-lying resonances decaying strongly in the 1s3l ionic states. Excitation from excited atomic states allow us to access autoionizing states of opposite parity from those achievable

from the ground state. Also, starting from excited atoms allows us to sample larger regions of space in the atom than for atoms in the ground state, providing a severe test of many-body calculations.

6) We have discovered the existence of several Rydberg series among the many hollow states. Using the multi-quantum defect theory, we have determined the quantum defect for three of them, again in excellent agreement with R-matrix calculations.[10]

7) We have succeeded to observe the first doubly hollow state, i. e., the lowest energy triply excited state with empty K- and L-shells. In the one electron notation, this state has a $3s^2 3p\ ^2P$ configuration. We measured the energy and the width of this state, in good agreement with the results of the Saddle-point and R-matrix calculations.[11]

8) One very important feature is also the angular dependence of autoionization which can reveal the symmetry of the hollow excited states. This is important to determine the identification and the mixing of the hollow atomic states. We have measured the first angle-resolved photoelectron data for two hollow states, namely the $2s^2 2p\ ^2P$ and $2p^3\ ^2P$ triply excited states in the one-electron notation.[7]

9) Above 160 eV photon energy and up to the triple ionization limit at 210 eV, we have made some exploratory experiments, which revealed many interesting new features that have never been seen before and are worthwhile to be investigated in more details.

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